

Title: Catalytic Gasification of Coal Using Eutectic Salt Mixtures

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ABSTRACT

The Gas Research Institute (GRI) estimates that by the year 2010, 40% or more of U.S. gas supply will be provided by supplements including substitute natural gas (SNG) from coal. These supplements must be cost competitive with other energy sources. Large-scale commercial plants to produce SNG from coal will need to be constructed around the turn of the century to meet these projected demands. Currently, proven so-called first generation technologies for coal gasification include moving-bed Lurgi Pressure Gasification Process, Entrained-Bed Koppers-Totzek Process, and the Fluidized-Bed Winkler Process. The most suitable for large-scale SNG production is the Lurgi Process at Sasol II and III and at the Great Plains Coal Gasification Plant. The relatively newer technologies that have the potential for SNG manufacture include KBW (Westinghouse) Ash Agglomerating Fluidized-Bed, U-Gas Ash Agglomerating Fluidized-Bed, British Gas Corporation/Lurgi Slagging Gasifier, Texaco Moving-Bed Gasifier, and Dow and Shell Gasification Processes. These processes are at various stages of development, ranging from demonstration units to commercial demonstration units. However, the relatively newer technologies have several disadvantages such as high severities of gasification conditions, low methane production, high oxygen consumption, inability to handle caking coals, and unattractive economics.

The objectives of this study are to: identify appropriate eutectic salt mixture catalysts for coal gasification; assess agglomeration tendency of catalyzed coal; evaluate various catalyst impregnation techniques to improve initial catalyst dispersion; evaluate effects of major process variables (such as temperature, system pressure, etc.) on coal gasification; evaluate the recovery, regeneration and recycle of the spent catalysts; and conduct an analysis and modeling of the gasification process to provide better understanding of the fundamental mechanisms and kinetics of the process.

A review of the literature was completed in the first six months of the project. The catalysts which have been used for gasification can be roughly classified under the following five groups: alkali metal salts; alkaline earth metal oxides and salts; mineral substances or ash in coal; transition metals and their oxides

and salts; and eutectic salt mixtures. Studies involving the use of gasification catalysts have been conducted. However, most of the studies focused on the application of individual catalysts. Only two publications have reported the study of gasification of coal char in CO₂ and steam catalyzed by eutectic salt mixture catalysts. By using the eutectic mixtures of salts that show good activity as individual compounds, the gasification temperature can be reduced possibly with still better activity and gasification rates due to improved dispersion of the molten catalyst on the coal particles. For similar metal/carbon atomic ratios, eutectic catalysts were found to be consistently more active than their respective single salts. But the exact roles that the eutectic salt mixtures play in these are not well understood and details of the mechanisms remain unclear. The effects of the surface property of coals and the application methods of eutectic salt mixture catalysts with coal chars on the reactivity of gasification will be studied.

Based on our preliminary evaluation of the literature, a ternary eutectic salt mixture consisting of Li-Na- and K- carbonates has been identified to have the potential as gasification catalyst. To verify the literature reported, melting points for various compositions consisting of these three salts and the temperature range over which the mixture remained molten were determined in the lab. For mixtures with different concentrations of the three salts, the temperatures at which the mixtures were found to be in complete molten state were recorded. The results are as follows:

Li ₂ CO ₃ (Gm)	Na ₂ CO ₃ (Gm)	K ₂ CO ₃ (Gm)	Temperature Range (F)
0.64	0.66	0.70	695-700
0.50	0.66	0.70	840-860
0.40	0.66	0.70	900-910
0.30	0.66	0.70	1000-1010

As shown above, by increasing the amount of Li₂CO₃, the melting temperature range was reduced significantly. In the literature, the eutectic mixtures of Li- Na- and K-carbonates are claimed to have a lower activation energy than that of K₂CO₃ alone and they remain molten at a lower temperature than pure K₂CO₃. The slow increase in the gasification rates with eutectics reported in the literature is believed to be due to a gradual penetration of the coals and coal char particles by the molten and viscous catalyst phase. The even spreading of the salt phase seems to increase the overall carbon conversion rate.

In the next reporting period, a number of eutectic salts and methods of their application on the coal will be identified and tested. The performance of the catalysts on the gasification of Illinois #6 coal will be evaluated through TGA and bench-scale fixed bed reactor studies. All chemicals and accessories, including the Illinois #6 coal, have been ordered.